

Searches for neutrinoless resonant double electron captures at LNGS

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Abstract. Several experiments were performed during last years at underground (3600 m w.e.) Laboratori Nazionali del Gran Sasso (LNGS) of the INFN (Italy) to search for resonant $2\varepsilon 0\nu$ captures in ^{96}Ru , ^{106}Cd , ^{136}Ce , ^{156}Dy , ^{158}Dy , ^{180}W , ^{184}Os , ^{190}Pt with the help of HP Ge semiconductor detectors, and ZnWO_4 and $^{106}\text{CdWO}_4$ crystal scintillators. No evidence for $r\text{-}2\varepsilon 0\nu$ decays was found, and only $T_{1/2}$ limits were established in the range of $10^{14} - 10^{21}$ yr.

1. Introduction

Resonant nuclear reactions occur sometimes in nature. Probably the most prominent example of such a phenomenon is a triple α reaction. There are no stable nuclei with $A = 5$ and 8. In these circumstances, the triple α reaction plays a critical role for nucleosynthesis of heavier elements, when at first two α particles create ^8Be nucleus (with very short $T_{1/2} \simeq 10^{-16}$ s) and one more α particle joins to create stable ^{12}C . Cross-section of the $^8\text{Be} + \alpha$ reaction is not big enough to explain abundances quantitatively. In 1953, F. Hoyle supposed [1] that

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^{12}C has an excited level at 7.68 MeV, and this should result in resonant enhancement of the cross-section by orders of magnitude. This level was searched for and immediately observed by experimentalists at 7.68 ± 0.03 MeV [2]. It seems, that our world and all of us exist because of resonant enhancement of this nuclear reaction.

Resonant enhancement is expected also for neutrinoless double electron capture ($2\varepsilon 0\nu$) $e_1 + e_2 + (A, Z) \rightarrow (A, Z - 2)$ in case of mass degeneracy of an initial and final (excited) nuclei:

$$Q = Q_{2\beta} - E_{b1} - E_{b2} = E_{exc},$$

where $Q_{2\beta} = \Delta M_a$ is the atomic mass difference between an initial and final nuclei.

The possibility of such resonant process ($r\text{-}2\varepsilon 0\nu$) was discussed long ago [3], where an enhancement of the rate by a few orders of magnitude was predicted for perfect energy coincidence between the released energy and the energy of an excited state ($\simeq 10$ eV). In this case the expected half lives could be as low as $\simeq 10^{24}$ yr, and the corresponding experiments even could compete with searches for neutrinoless $2\beta^-$ decay in sensitivity to the neutrino mass. Theoretical aspects are discussed in recent papers [4].

The first experimental limit on the $r\text{-}2\varepsilon 0\nu$ process, to our knowledge, was obtained for ^{106}Cd $2K$ capture to ^{106}Pd level with $E_{exc} = 2741$ keV: $T_{1/2} > 3.0 \times 10^{19}$ yr at 90% C.L. [5]. It was achieved in underground measurements at LNGS of 154 g sample of Cd enriched in ^{106}Cd to 68% with two low background NaI(Tl) scintillators working in coincidence. This enriched ^{106}Cd isotope was used to grow $^{106}\text{CdWO}_4$ crystal scintillator [6] for our further investigations of 2β processes in Cd and W.

Below, summary of our searches for $r\text{-}2\varepsilon 0\nu$ captures in different nuclides is given; the experiments were performed mainly during last two years.

2. Experiments

Different approaches were used in the experiments: sharp peaks that correspond to γ quanta emitted in deexcitation of $(A, Z - 2)^*$ nuclei were searched for $r\text{-}2\varepsilon 0\nu$ processes in ^{96}Ru , ^{136}Ce , ^{156}Dy , ^{158}Dy , ^{184}Os , ^{190}Pt with the help of HP Ge detectors, while ZnWO_4 and $^{106}\text{CdWO}_4$ crystal scintillators were applied in investigations of ^{180}W and ^{106}Cd , respectively. Response functions and efficiencies were calculated with the GEANT4 simulation tool [7]; initial kinematics of particles emitted in decays was generated with the DECAY0 event generator [8].

Natural isotopic abundance of ^{96}Ru is $\delta = 5.54\%$. Ru sample with mass of 473 g was measured with HP Ge detector of 468 cm^3 during 158 h at the first step; then data were collected also by a set-up with four HP Ge detectors ($\simeq 225$ cm^3 each) during 1176 h [9]. $T_{1/2}$ limits were established at the first time for $r\text{-}2\varepsilon 0\nu$ in ^{96}Ru at the level of 10^{19} yr (see Table 1). Quite strong pollution of the Ru sample by ^{40}K at 3.4 Bq/kg was found [9]. Additional purification allowed to suppress ^{40}K by one order of magnitude, and new data are under collection with the purified Ru sample with mass of $\simeq 0.7$ kg.

For investigations of ^{106}Cd , scintillating crystal $^{106}\text{CdWO}_4$ was developed [6] with mass of 215 g and 66.4% enrichment in ^{106}Cd (while natural δ is 1.25%). The energy resolution of the detector is 10% at 662 keV. After 6590 h of data taking, new improved half life limits on the $r\text{-}2\varepsilon 0\nu$ processes were established at the level of up to 10^{21} yr [10], competitive or better than those set in the TGV experiment [11].

A sample of CeCl_3 crystal (6.9 g) was used to search for $r\text{-}2\varepsilon 0\nu$ in ^{136}Ce with HP Ge 244 cm^3 during 1280 h [12]. $T_{1/2}$ limits were obtained for this nuclide at the first time, but the small mass and poor natural abundance (0.185%) allowed to obtain the $T_{1/2}$ values on the level of only 10^{15} yr.

First searches for $r\text{-}2\varepsilon 0\nu$ decays of ^{156}Dy and ^{158}Dy were performed with Dy_2O_3 sample with mass of 322 g (99.98% purity grade) and HP Ge detector 244 cm^3 during 2512 h. Once more,

$T_{1/2}$ limits were not very big (around 10^{16} yr) because of low natural abundance of ^{156}Dy and ^{158}Dy isotopes (0.056% and 0.095%, respectively) [13].

Several ZnWO_4 crystal scintillators with mass up to 699 g were used to search for $r\text{-}2\varepsilon 0\nu$ decay of ^{180}W to the ground state of ^{180}Hf . After near 19,000 h of measurements, the $T_{1/2}$ limit was set as 1.3×10^{18} yr [14].

For investigations of ^{184}Os , sample of natural Os with mass of 172.5 g and purity grade of $> 99.999\%$ (purified in the Kharkiv Institute of Physics and Technology, Ukraine; probably this is the most pure Os in the world) is under measurements now with HP Ge 468 cm^3 . The results are expected soon.

Natural platinum sample with mass of 42.5 g was measured with HP Ge detector 468 cm^3 during 1815 h. The ^{190}Pt isotope has quite low natural abundance of 0.014% that leads to not very high $T_{1/2}$ limit for $r\text{-}2\varepsilon 0\nu$ decay of 2.9×10^{16} yr [16]. As by-product of these measurements, also α decay of ^{190}Pt to the first excited level ($E_{exc} = 137.2$ keV) of ^{186}Os was observed at the first time; corresponding half life is $T_{1/2} = 2.6^{+0.4}_{-0.3}(\text{stat.}) \pm 0.6(\text{syst.}) \times 10^{14}$ yr [17].

All the obtained results are summarized in Table 1 where also the expected energy releases Q are compared with the energies of excited levels of the final nuclei. It should be noted that the Q values used in our papers on Ru, Ce, Dy were based on the atomic masses [18] usually known with accuracy of a few keV. Very recently many new, much more accurate results on the atomic masses appeared, motivated also by searches for perfect candidate for $r\text{-}2\varepsilon 0\nu$ process. The new values could be significantly shifted from the old ones sometimes removing an isotope from the list of perspective $r\text{-}2\varepsilon 0\nu$ candidates. In particular, the old $Q_{2\beta}$ values [18] versus the new ones are the following: for ^{96}Ru 2718 ± 8 vs. 2714.51 ± 0.13 keV [19], for ^{106}Cd 2770 ± 7 vs. 2775.39 ± 0.10 keV [20], for ^{136}Ce 2419 ± 13 vs. 2378.53 ± 0.27 keV [21], for ^{156}Dy 2012 ± 6 vs. 2005.95 ± 0.10 keV [22]. However, one should remember also about possibility that some new excited levels in daughter nuclei could be discovered with energies and J^π values well fitted for the new atomic masses.

3. Conclusions

Resonant $2\varepsilon 0\nu$ captures in ^{96}Ru , ^{106}Cd , ^{136}Ce , $^{156,158}\text{Dy}$, ^{180}W , ^{190}Pt were searched for with HP Ge spectrometry and with scintillating crystals ZnWO_4 and $^{106}\text{CdWO}_4$. Only $T_{1/2}$ limits were established from 3.0×10^{14} to 9.6×10^{20} yr. These values are mostly the best today for these nuclides, sometimes better than the previous ones by few orders of magnitude, and sometimes they were obtained at the first time. However, they are still orders of magnitude worse than those predicted by theory [3, 4]. An “excellent” candidate for $r\text{-}2\varepsilon 0\nu$ is still not found.

In searches for $r\text{-}2\varepsilon 0\nu$ captures, exact knowledge of $Q_{2\beta}$ values and J^π properties of excited levels is needed. Significant progress was reached recently for measurements of $Q_{2\beta}$ for many 2β nuclides but we still need more accurate information for other isotopes.

Interesting by-products sometimes happen, like the first observation of α decay $^{190}\text{Pt} \rightarrow ^{186}\text{Os}^*$ ($E_{exc} = 137.2$ keV) with $T_{1/2} = 2.6 \times 10^{14}$ yr [17]. Experiments to search for resonant $2\varepsilon 0\nu$ captures in ^{96}Ru , ^{106}Cd and ^{184}Os are in progress at LNGS.

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Table 1. Summary of results on $r\text{-}2\varepsilon 0\nu$ captures obtained in our experiments. $T_{1/2}$ limits (yr) correspond to 90% C.L. The Q values were calculated with the new atomic masses for ^{96}Ru [19], ^{106}Cd [20], ^{136}Ce [21], ^{156}Dy [22]; for other nuclei masses of [18] were used.

Transition		Q (keV)	E_{exc} (keV), J^π	Lim $T_{1/2}$	Method	Year
$^{96}\text{Ru} \rightarrow ^{96}\text{Mo}$	KL_1	2691.6(1)	2700.2(1), 2^+	2.2×10^{19}	HP Ge	2009 [9]
	$2L_1$	2708.8(1)	2712.7(1)	5.1×10^{19}		
$^{106}\text{Cd} \rightarrow ^{106}\text{Pd}$	$2K$	2726.7(1)	2717.6(2)	4.3×10^{20}	$^{106}\text{CdWO}_4$	2011 [10]
	KL_1	2747.4(1)	2741.0(5), 4^+	9.5×10^{20}		
	KL_3	2747.9(1)	2748.2(4), $2, 3^-$	4.3×10^{20}		
$^{136}\text{Ce} \rightarrow ^{136}\text{Ba}$	$2L_1$	2366.6(3)	2392.1(6), $(1^+, 2^+)$	2.4×10^{15}	HP Ge	2009 [12]
	$2L_1$	2366.6(3)	2399.9(1), $(1^+, 2^+)$	4.1×10^{15}		
$^{156}\text{Dy} \rightarrow ^{156}\text{Gd}$	$2K$	1905.5(1)	1914.8(1), 2^+	1.1×10^{16}	HP Ge	2011 [13]
	KL_1	1947.3(1)	1946.4(1), 1^-	9.6×10^{15}		
	KL_1	1947.3(1)	1952.4(1), 0^-	2.6×10^{16}		
	$2L_1$	1989.2(1)	1988.5(2), 0^+	1.9×10^{16}		
$^{158}\text{Dy} \rightarrow ^{158}\text{Gd}$	$2L_3$	1991.5(1)	2003.7(1), 2^+	3.0×10^{14}	HP Ge	2011 [13]
	$2L_1$	268(3)	262, 4^+	3.2×10^{16}		
	$2K$	13(4)	g.s., 0^+	1.3×10^{18}		
	$2K$	1312(1)	1322, $(0)^+$	—		
$^{180}\text{W} \rightarrow ^{180}\text{Hf}$	KL_1	1370(1)	1360, (4^+)	—	HP Ge	2011 [15]
	$2L_1$	1427(1)	1425, $(3)^+$	—		
	$2L_1$	1427(1)	1431, 2^+	—		
	$2L_1$	1427(1)	1431, 2^+	—		
$^{190}\text{Pt} \rightarrow ^{190}\text{Os}$	$2M$	1378(6)	1382, $(0, 1, 2)^+$	2.9×10^{16}	HP Ge	2011 [16]

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